

Non-Newtonian Fluid Mechanics

(Molecular Mechanisms)

*March 1, 1965 - Aug. 31, 1965*  
Contract Number: NsG 667

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Objective: The purpose of this research is to determine and study the molecular mechanisms responsible for Newtonian and non-Newtonian viscosity in high-polymer melts and solutions.

Background: This work is part of a larger NASA sponsored research program to study non-Newtonian fluid properties and fluid mechanics. Although this report covers the first year of the existence of NsG 667 our research group was formed in January 1965 and has been active for only about 8 months. Much of this time has been spent in setting up, equipping, and organizing two new polymer research laboratories. We have received support from three sources to study the broad area of molecular relaxation and flow in polymer melts and solutions. The largest support is from NsG 667 with somewhat less from the University of Pennsylvania (administering ARPA funds) and an NSF research initiation grant. We expect our research, within this broad area, to evolve into three clearly defined projects within 12 to 18 months. During these early stages of the research, however, we find it difficult to assign the specific origin of support for any particular piece of work since our activity has been largely restricted to: (1) selection, construction, and testing steady state and dynamic viscometers, (2) bringing the graduate students up to a sufficient level of experimental and/or theoretical sophistication that they can do relatively independent work, (3) rather general computer programming, (4) developing auxiliary experimental techniques such as preparation of monodisperse polymer, preparation of uniform polymer solutions (of high viscosity) and recovery of polymer from solution. Consequently we will describe below our total progress and activity in the field of relaxation and flow in high-polymer systems.

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| FACILITY FORM 802 | N 66-80378                    |             |
|                   | (ACCESSION NUMBER)            | (THRU)      |
|                   | 3                             | <i>None</i> |
|                   | (PAGES)                       | (CODE)      |
|                   | CR-68283                      |             |
|                   | (NASA CR OR TMX OR AD NUMBER) | (CATEGORY)  |

Progress, Activity and Immediate Plans: A Haake Rotovisco viscometer was obtained for our steady state viscosity measurements. It is equipped with cup-and-bob and cone-and-plate attachments. Our tests show the instrument capable of measuring viscosity as a function of shear rate on our polystyrene solutions from 15% to 50% polymer. This covers the concentration range of most interest to us at present, but we plan to get a second cup-and-bob attachment to enable us to go to much lower concentrations.

The instrument to measure dynamic viscosity is nearly completed. The "heart" of the dynamic viscometer is the Haake Rotovisco. The Rotovisco is driven in a sinusoidal mode with a multi-frequency torsional pendulum. The resonance frequency of the pendulum is designed to be varied between 0.05 and 1000 cycles per sec. by varying the torsion bar and inertial elements. The amplitude of the pendulum is measured with an optical-electronic transducer system. Depending on the frequency, either the amplitude of the oscillation or its envelope is recorded. Motion of the pendulum is also monitored with an oscilloscope. The period of the vibration will be measured with a Hewlett-Packard counter (soon to be ordered). By measuring resonance frequency and log-decrement of the pendulum-viscometer system with and without sample, we can determine the dynamic viscosity of the polymer melt or solution.

The torsion pendulum operates well between 0.1 and 100 cycles per sec., but mechanical instability makes measurements impossible at higher frequencies. We have redesigned the system incorporating a nearly frictionless air bearing. If our calculations are correct, lateral motion of the inertial element will be held to  $\pm 3 \times 10^{-4}$  inch. After many modifications we have developed an optical-electronic system that operates adequately. We expect to improve our signal to noise ratios, however, by further electrical and light shielding.

The instrument constants of the dynamic viscometer are now being obtained by calibration with non-viscoelastic (and Newtonian) mineral oil. After calibration we will make pilot runs, over our now limited frequency range, using solutions of inexpensive heterodisperse polystyrene. After determining the most appropriate concentration and temperature ranges, and after our new air bearing has been installed, we will proceed with measurements on solutions of our rather precious samples of monodisperse polymer. (generous gifts from Dow Chemical Co. and the Mellon Institute).

To date all our attempts to prepare monodisperse polystyrene have failed. We have apparently not yet been able to develop the art of maintaining the ultra-purity required of all the monomer and solvents. Although we still have hopes of making our own monodisperse polymer, this phase of our work is no longer critical since it is now available commercially at about ten dollars per twelve grams.

Since much of our work will be with solutions of viscosities in excess of 100 Poise, forming homogeneous solutions becomes a major problem. We have designed a special mixer which we feel will do the job. It is currently under construction.

In anticipation of obtaining our first steady state and dynamic viscosities, we have written a computer program to calculate viscosities and a number of viscoelastic functions (creep compliance, stress relaxation, dynamic modulus, dynamic compliance, and loss factor). The program as it now exists will calculate all of the above functions from the Rouse-Bueche relaxation theory, but it can be easily modified to do the calculations starting from any discrete distribution of relaxation times.

Date: October 25, 1965